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Water-air-CO_2-flux changes after damming rivers loaded with suspended basaltic particles

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ABSTRACT

Contributions of CO_2 emissions from reservoirs to the atmosphere are continuously increasing with rising energy demand. Therefore, it is important to quantify the emissions and define the rate determining mechanism of CO_2 fluxes in man-made reservoirs. Here we present results from two reservoirs in Iceland over a total time span of 16 years. The partial pressure of CO_2 within the Hálslón reservoir, fed by glacier meltwater loaded with suspended basaltic particles, was considerably less than the CO_2 pressure of the atmosphere during the years 2008–2013. The specific CO_2 uptake from the atmosphere into Hálslón was estimated at 121 ± 67.9 g_{CO2} m^{-2} yr during the 6 months ice-free period or 5000 t annually. The uptake rate was governed by the CO_2 gradient across the water-air-interphase and windspeed but less by temperature. However, temperature will affect water-rock interactions and sub-zero temperature can result in ice cover, terminating water-air interactions. Atmospheric CO_2 concentration dictates the maximum upper limit of the CO_2 influx rate at fixed wind speed.

The downstream mixing of Hálslón reservoir water with the CO_2 emitting Lagarfljót reservoir lowered the CO_2 emissions from Lagarfljót from 5335 t CO_2 yr^{-1} to 1670 t CO_2 yr^{-1} after the damming. This study shows that dissolution of basalt in glacier melt waters leads to direct CO_2 uptake from the atmosphere, which can potentially be utilised for future carbon removal from the atmosphere.

1. Introduction

The present 8 Gt CO_2 yr^{-1} release of CO_2 from Earth's terrestrial surface waters to the atmosphere is a major contributor to the short-term carbon cycle (Raymond et al., 2013). This flux from surface waters is much larger than the 2 Gt CO_2 yr^{-1} degassing from the Earth's crust and the 0.5 Gt CO_2 yr^{-1} combined CO_2 drawdown from the atmosphere by weathering of silicates and carbonates on continents (Gaillardet et al., 1999; Hartmann et al., 2009). According to Raymond et al. (2013) the global emissions from inland waters can be divided into streams and rivers (6.6 Gt CO_2 yr^{-1}) and lakes and reservoirs (1.2 Gt CO_2 yr^{-1}). Changes in the course of rivers, e.g. by damming, can therefore significantly affect CO_2 emissions. Currently, emissions from hydropower reservoirs are estimated at 0.38 Gt $CO_{2\,eq}$ yr^{-1} globally (Li and He, 2022).

Within the present study we quantify the water-air-CO_2 exchange through the surface of the Hálslón reservoir constructed in 2003–2007, located at the northeastern edge of the Vatnajökull glacier, Iceland, and the water-air-CO_2 exchange before and after the modification of the Lagarfljót reservoir downstream (Fig. 1). This field site in NE-Iceland represents the extreme case where the reservoir waters are poor in suspended particulate organic matter but loaded with reactive basaltic particles, capable of lowering the partial pressure of CO_2 in these waters via water-rock interactions, resulting in direct uptake of CO_2 from the atmosphere.

Here we report the water chemistry and suspended material composition of the new Hálslón reservoir in NE-Iceland, the outflow from the associated Kárahnjúkar power plant, and the downstream Lagarfljót reservoir waters before and after mixing with the Hálslón reservoir waters. Secondly, the measured air temperature and wind speed distribution at the Kárahnjúkar dam and the Lagarfljót reservoir at Egilsstaðir, before and after commission of the Kárahnjúkar dam, are presented. We calculated the in situ CO_2 partial pressure values of the reservoir waters and compared them to the measured daily average CO_2 concentration in the atmosphere at the National Oceanic and Atmospheric Administration (NOAA) Stórhöfði South Iceland monitoring

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station. These were then used to calculate the CO_2-fluxes between these reservoir waters and the atmosphere during the sampling periods 1998–2003, and 2008–2013. Finally, we present a comparison of the water-air-fluxes with the combined dissolved inorganic carbon (DIC) river fluxes from these reservoirs to the ocean.

The measurements of discharge and chemical composition of the river waters and their suspended material before and after the commission of the Kárahnjúkar power plant revealed a doubling of the river discharge through the Lagarfljót reservoir at the Lagarfoss dam and significant changes in the associated river fluxes (Eiriksdottir et al., 2008, 2013, 2014; 2015; 2017; Gislason et al., 2006, 2009; Louvat et al., 2008). Here we used this existing data to quantify the CO_2 water-air fluxes using additional climatological data and established modelling approaches to quantify the direct water-air-CO_2 fluxes, and to illuminate the rate controlling factors.

2. Methods

2.1. Field site

The location of the studied field site affected by the damming of the Jökulsá á Dal glacial river in Eastern Iceland is shown in Fig. 1. Related to the construction of the Kárahnjúkar power plant in 2003–2007, potentially affected nearby rivers were monitored from 1998 to 2003, and after the damming from 2008 to 2013. The newly created Hálslón reservoir reaches a geographic surface area of up to 58.3 km^2 resulting in a total volume of 2.2 km^3 (Leifsson et al., 2009). The reservoir is within 25 km of the northern edge of the Vatnajökull glacier (Leifsson et al., 2009; Gunnarsson et al., 2014). In May each year (Fig. S1), when the reservoir is at its lowest filling level, the surface area is about 17 km^2 (Leifsson et al., 2009). If the Hálslón reservoir reaches its minimum operational level, the power plant uses water from two other reservoirs, Kelduárlón (Fig. 1) and Ufsarlón, situated in the catchment of the Jökulsá í Fljótsdal river.

A headrace tunnel channels the water from the reservoirs to the power plant (see Fig. 1). Afterwards, this water is then released into the Jökulsá í Fljótsdal river (normal water level ~ 25 m a.m.s.l., Leifsson et al., 2009) and eventually into the Lagarfljót reservoir. Its average water level at the Lagarfoss dam was at 19.9 m a.m.s.l. before the operation of the Kárahnjúkar dam (Axelsson, 2012). The Lagarfljót reservoir is a 35 km long lake with a geographic surface area of 53 km^2, a maximum depth of 112 m, and a volume of 2.7 km^3 (Hallgrímsson, 2005). The average discharge through the Kárahnjúkar headrace tunnel is 110 m^3 s^{-1} (Landsvirkjun, 2009), causing the average discharge from the Lagarfljót at Lagarfoss to double after the commissioning of the Kárahnjúkar power plant, November 2nd, 2007. Consequently, the water residence time in the Lagarfljót reservoir was halved, from one year to six months (Tómasson and Hardardóttir, 2001).

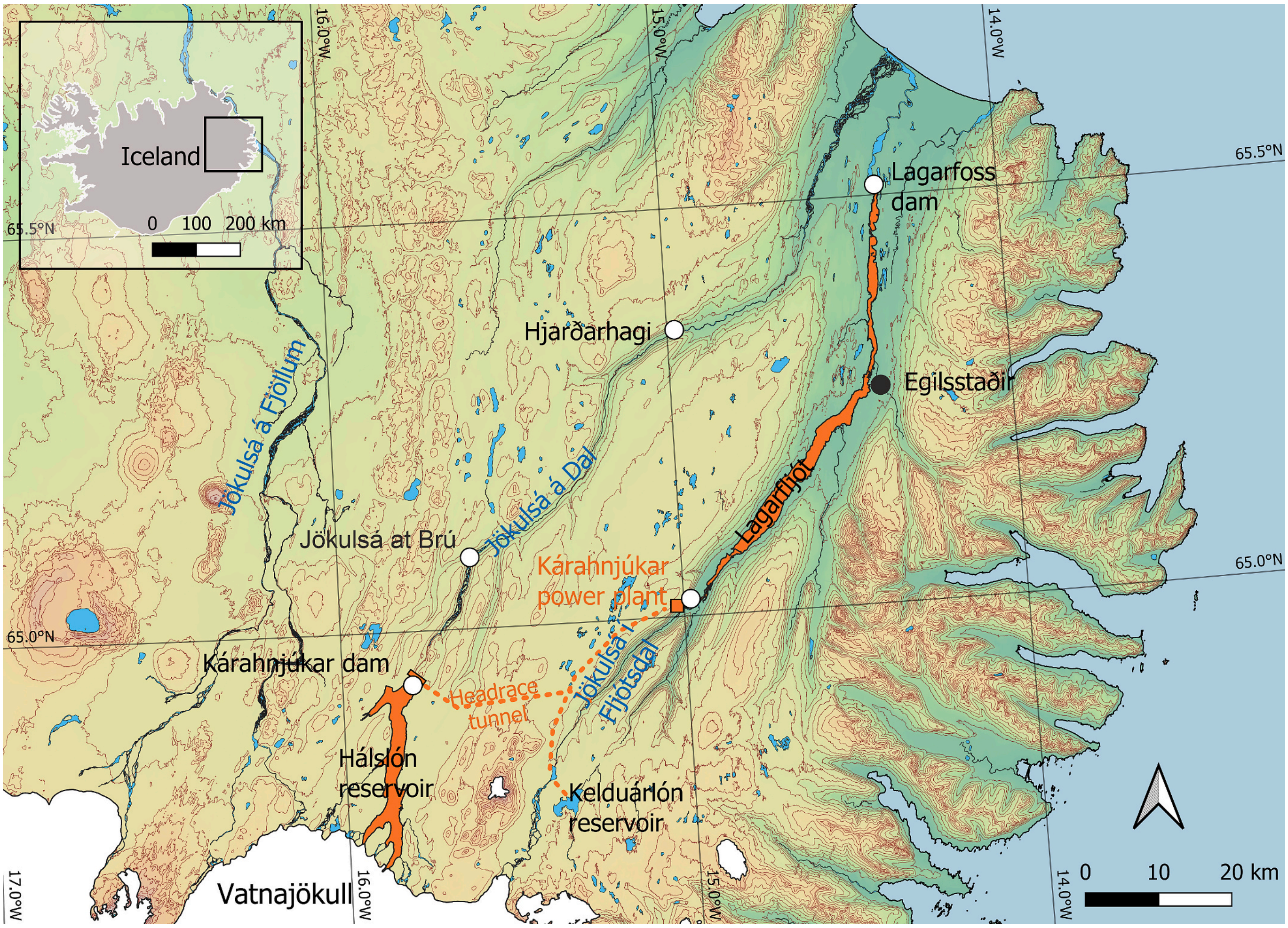

Fig. 1. Map showing the location of the two river catchments affected by the damming of the Jökulsá á Dal glacial river in Eastern Iceland. Displayed are the location of the sampling sites (white filled circles). In orange colour are the Hálslón reservoir created by the Kárahnjúkar dam, the headrace tunnels (orange dashed curves), the power plant (orange square), and the Lagarfljót reservoir downstream from the power plant extending all the way down to the Lagarfoss dam. The Vatnajökull glacier is shown in white in the lower left corner. Weather stations are close to the Kárahnjúkar dam and Egilsstaðir (black filled circle). Map data from the National Land Survey of Iceland and Landsvirkjun, National Power Company of Iceland.

These river catchments are partially glaciated. Before the construction of the Kárahnjúkar dam glaciers covered 43 % and 6 % of the catchments at Jökulsá á Dal and Lagarfljót, respectively, above the monitoring sites in Hjarðarhagi and Lagarfoss (Fig. 1) (Kardjilov, 2008). The glacier cover of the Hálslón reservoir catchment is 78 % (Leifsson et al., 2009). Every year after the Hálslón reservoir fills up during late summer, and overflows via a spillway into the Jökulsá á Dal river channel for a few weeks, it dramatically changes the river discharge and the riverine concentration of suspended and dissolved material downstream of the reservoir (Eiriksdottir et al., 2017).

2.2. Water sampling and analysis

The collection and analysis of samples was previously described by Eiriksdottir et al. (2013, 2014, 2015, 2017). Samples were collected throughout the year to account for seasonal changes on the chemical compositions of the rivers and the outlet from the power plant (Tables S1–5 and S7). Additionally, 21 samples, given in Table S6, were collected from the Hálslón reservoir closest to its maximum depth at 64°56′24.24”N 15°47′32.46”W, at various depths in May 2008 through 80 cm thick ice (Fig. S3–4, Table S6) and by boat in August 2008 (Fig. S5). Further samples were collected in August 2009 from 5 m depth and in September as surface samples pulled in from the shore. One more surface sample was taken in August 2010. After that, samples were taken from a boat at various depths above the maximum depth of the reservoir at 64°56′29.28”N 15°47′40.56”W when water was overflowing the dam via the spillway in 2011, 2012 and 2013 (Fig. S5). All these reservoir samples were collected with a Niskin sampler and treated the same as the river samples. Simultaneously, detailed temperature profiles from the surface and down to about 130–160 m depth were measured in 2008, 2011, 2012 and 2013 (Fig. S5 and Table S6 in the Supplement). A more detailed sampling description can be found in the supplementary material and the above cited literature.

Water temperature profiles within the Hálslón reservoir water body were measured from a boat in August 2008 at the deepest part of the reservoir shown by the white filled circle in the Hálslón reservoir in Fig. 1, and again at the same location in September when the water level had reached the spillway at more than 625 m a.m.s.l. and yet again in 2011, 2012 and 2013 (Eiríksdóttir et al., 2014; Fig. S5).

2.3. Chemical speciation and CO_2 flux calculations

The CO_2 flux F (mol m^{-2} s^{-1}) across the air-water interface was calculated from the product of the gas transfer velocity k (m s^{-1}) and the CO_2 concentration difference (see eq. 1, Wanninkhof et al., 2009). We used PHREEQC interactive software version 3.4.0 (Parkhurst and Appelo, 1999) and the minteq.v4.dat database (Allison et al., 1991; U.S. Environmental Protection Agency, 1998) to determine the chemical speciation of the water samples and the in situ partial pressure (atm) of CO_2 in the well mixed bulk fluid pCO_{2w}. The partial pressure of the atmosphere pCO_{2atm} was derived from the data of the NOAA CO_2 monitoring station (Lan et al., 2023) in South Iceland.

$$F_{CO2} = k\,K_H(pCO_{2w} - pCO_{2atm}) \quad (1)$$

The transfer velocity *k* was estimated using the approach by Wanninkhof (1992), where k is parameterized to the temperature-dependent nondimensional Schmidt's number Sc, which is equal to 600 for CO_2 at 20 °C in fresh water (Jähne et al., 1987).

$$k = k_{600}(Sc_{CO2}/600)^n \quad (2)$$

where k_{600} is the gas transfer velocity normalized to Sc of CO_2 at 20 °C with $n = 2/3$ for wind speeds U less than 3.6 m s^{-1} and $n = 1/2$ for wind speeds above 3.6 m s^{-1} (Jähne et al., 1987).

The Schmidt's number Sc_{CO2} was extrapolated to other water temperatures (Wanninkhof, 1992) using the monthly median air temperatures from nearby meteorological stations measured at 2 m above ground level (a.g.l.). For all calculations all temperatures at or below 0 °C were set to 0.1 °C, to avoid numerical problems.

k_{600} was calculated following Cole and Caraco (1998):

$$k_{600} = 2.07 + 0.215 \bullet U_{10}{}^{1.7} \quad (3)$$

together with the wind speeds measured at 10 m height a.g.l. from nearby meteorological stations.

2.4. Wind speed and air temperature

The wind speed and air temperature data are received from the data base of the Icelandic Meteorological Office (IMO) with a temporal resolution of 1 h. Data is obtained from two stations, Kárahnjúkar (No. 5933, 64°56′49.6”N 15°47′34.5”W, 639 m a.m.s.l.) by the Hálslón reservoir and Egilsstaðir airport (No. 4271, 65°16′29.0”N 14°24′23.3”W, 23.5 m a.m.s.l.) by the Lagarfljót reservoir (see Fig. 1). The wind velocity was measured by standard Young anemometers at 10 m a.g.l, with an accuracy of ±0.3 m s^{-1}. and the air temperature by Logan platinum resistance thermometers at 2 m a.g.l, with an accuracy of ±0.1 K. Data was retrieved from the Kárahnjúkar station for the time period June 2008 to September 2013, including the duration of the water monitoring period of the Hálslón and the Lagarfljót reservoirs, and from the Egilsstaðir airport weather station from November 1998 to November 2003 (before the deviation of the Jökulsá á Dal river into Lagarfljót) and from November 2007 to December 2013 (after the deviation).

2.5. Estimation of Ice cover

The determination of the ice cover of the reservoirs is done by visual assessment through web cameras operated by the Landsvirkjun power company. During spring and autumn when changes in the ice conditions are expected, web cameras are regular examined, and the conditions of the reservoirs are noted. Low visibility during weather events (snow/fog) can affect the estimation of the dates, leading to some additional uncertainty if the web cameras cannot capture the entire reservoir. Estimation of ice formation is more precise as the reservoirs usually freeze over all at once. The freezing and melting dates have an uncertainty of up to one week.

3. Results

3.1. Temperature and wind speed

The wind speed and temperature distribution measured at Kárahnjúkar and Egilsstaðir airport for each calendar month are provided in the Supplement Fig. S6 and Table S8. Generally, the highest wind speeds are observed during the winter months, while the lower wind speeds are present during summer when higher temperatures occur. During storm events, the wind speeds can exceed 30 m s^{-1}, but typically only lasting for a few hours. Wind speeds above 13 m s^{-1} were typically not observed for more than 24 consecutive hours. Therefore, this time span was used to estimate maximum fluxes from high wind speed events. The wind speeds measured at the Kárahnjúkar weather station are higher than that at the weather station at the Egilsstaðir airport, while the Kárahnjúkar temperatures are lower, due to the higher elevation. The air temperatures are similar during the two monitoring periods at the Egilsstaðir airport, but the wind speeds have declined slightly with time.

The air temperature at the Hálslón reservoir was higher during the August campaign than during those in September, and strong wind prevailed before and during the 2011, 2012 and 2013 campaigns. The water temperature from the surface down to ~50 m water depth was higher in August 2008 (~6 °C) than during September of the following years (only 3–4 °C, Table S6).

Additional temperature profiles were measured in Hálslón six times per year, from June to November 2009 to 2012, by the Landsvirkjun Power Company (Böðvarsdóttir et al., 2014). The highest measured temperatures are in late July to early August (6° to 8 °C), the lowest temperatures are measured in early June and late November (1 °C to 2.5 °C). The shallowest measurements reported were at 1 m depth in 2009–2011 and at 10 m depth in 2012. Hence, the temperature span of all the shallow waters (0–10 m depth, Figs. S4-S5) during the ice-free period, ranges from an average temperature of 4.5 ± 4 °C. This is in good agreement with the monthly median air temperature from the Kárahnjúkar weather station during the ice-free period (see Supplement Fig. S6). In the following, monthly mean air temperatures and wind speeds were used to calculate the monthly mean gas transfer.

3.2. *Ice cover*

Field observations show that the Hálslón reservoir was fully covered with ice from mid to end of November during the water monitoring period from 2009 to 2013 (see Table S10). Ice was fully melted by 1st to 12th of June during 2009 to 2012. No ice cover data is available for the Hálslón reservoir for the year 2008, but the reservoir was fully covered with ice May 19th, 2008, when the temperature profile was measured (Fig. S3–4) The reservoir was ice-free for about half a year in 2014–2019, from June to November–December (average 179 days).

For the study period of 1998 to 2013, the Lagarfljót reservoir as reported by Landsvirkjun was never fully frozen. Therefore, no ice-cover effect on the CO_2 fluxes were assumed for this reservoir.

3.3. *Chemical composition of water samples*

The chemical compositions of the studied rivers before and after the damming and the associated reservoirs are presented in the supplementary Tables S1-S7. Much of this data has been previously reported by Eiriksdottir et al., 2015, 2017. Here we have added the composition of the Lagarfljót reservoir waters and particles before the installation of the Kárahnjúkar dam (Table S4). Furthermore, to all tables we have added the mass percent of organic carbon in the suspended organic and inorganic particles, the molar precent of dissolved organic carbon (DOC) in the total dissolved carbon (TDC = DIC + DOC) in each sample, the C/N molar ratio of the organic particles, the in situ pH, the in situ pCO_{2w} and $CO_{2(aq)}$, which is the in situ concentration of dissolved CO_2 in the water. In situ refers to the calculated value at the measured water temperature at the time of sampling.

3.4. *CO_2 partial pressures and fluxes*

3.4.1. *Lagarfljót reservoir*

The calculated pCO_{2w} for the Lagarfljót reservoir are shown in Fig. 2 and in the Supplementary Information in Tables S4, S5 and S9. Before the damming (1998–2003) the average pCO_{2w} was 507 µatm. The pCO_{2w} values were mostly at or above the partial pressure of the atmosphere, pCO_{2atm}, at the time of sampling (avg. 367 µatm, calculated from Lan et al., 2023). After the construction of the Kárahnjúkar power plant in 2007 and the ensuing change in the Lagarfljót reservoir's inflow, the pCO_{2w} values decrease to an average of 444 µatm, while the atmospheric pCO_{2atm} values increased due to continuous rise in man-made global CO_2 emissions (avg. 393 µatm, calculated from Lan et al., 2023). Overall, the partial pressure values were above the atmospheric reference in both cases, indicating a CO_2 flux from the reservoir to the atmosphere, but the difference (pCO_{2w}-pCO_{2atm}) became smaller (from 140 µatm to 52 µatm) decreasing the CO_2 flux to the atmosphere after the construction of the Kárahnjúkar power plant (see Fig. 2 and Table S9).

The CO_2 fluxes from the Lagarfljót reservoir before (1998–2003) and after the damming of the Hálslón reservoir (2007–2013) were calculated from the difference of pCO_{2atm} and pCO_{2w}, using the pCO_{2atm} derived from the average pCO_2 values (367 µatm and 393 µatm) over the measuring period from the NOAA station and the average pCO_{2w} of all measured water samples before (507 µatm) and after the damming (444 µatm) respectively. Flux calculations were done for each month, using median monthly air temperatures and median monthly wind speeds, and a constant surface area of 53 km^2. Error estimates were done using the upper and lower quartile of the median monthly temperatures and wind speeds as (see Fig. 3). Additionally, we estimated potential high fluxes during short time (24 h) high wind speed events based on the highest hourly outliers of wind speed measured for each month (see Supplementary Information and results in Table S9). The average annual flux was calculated as a sum of the monthly fluxes, with additional information from the error estimates. The ice cover during the winter is assumed to prevent any gas exchange with the atmosphere, reducing

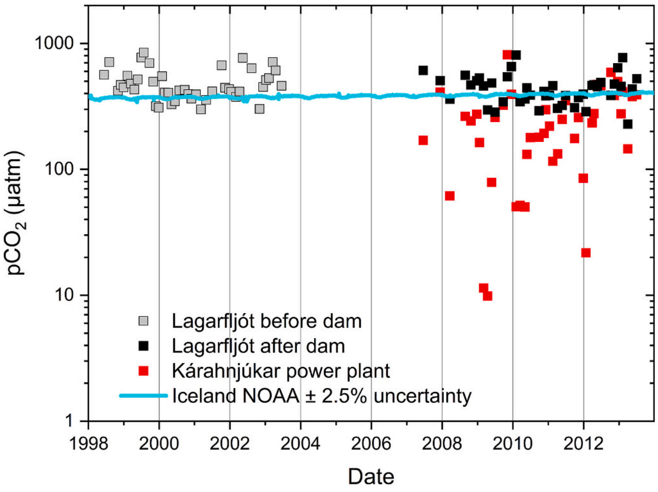

Fig. 2. Time evolution, before (grey filled squares) and after the erection of the Kárahnjúkar dam (black filled squares), of the in situ partial pressure of CO_2 in the Lagarfljót reservoir water at the Lagarfoss dam and in the outlet from the Kárahnjúkar power plant (red filled squares) sampled close to the power plant (Fig. 1). The light blue curve represents the CO_2 partial pressure in the atmosphere at the NOAA Stórhöfði south Iceland CO_2 monitoring station (Lan et al., 2023). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

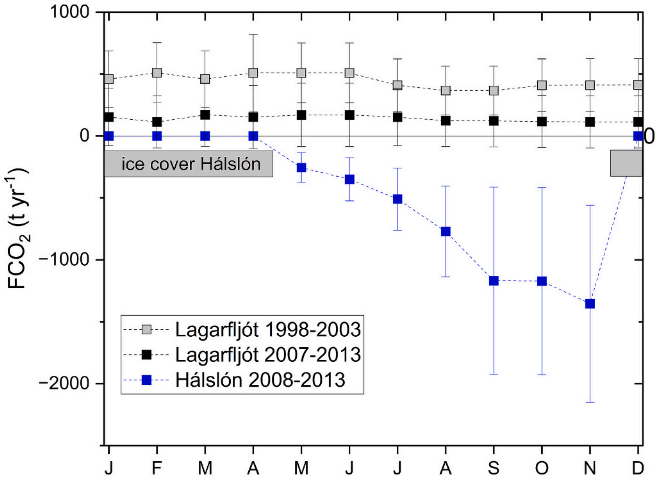

Fig. 3. Monthly mean CO_2 fluxes of the three datasets: Lagarfljót reservoir before (1998–2003) and after (2007–2013) the construction of the Kárahnjúkar power plant, and the Hálslón reservoir during 2008–2013. Error range based on monthly temperature and windspeed variations. Note, each year the Hálslón reservoir is covered with ice approximately from November to May resulting in termination of the CO_2 fluxes.

potential fluxes during these months. But as the Lagarfljót reservoir was not reported to fully freeze during the winter months, no ice cover has been assumed, therefore the fluxes represent maximum values without any effects of blockage by ice.

The annual CO_2 evasion from the Lagarfljót reservoir was estimated to be around **5335 ± 2736 t** before the construction of Kárahnjúkar power plant. After the dam construction the annual flux was less than one third, **1670 ± 2741 t,** for the observation period of 2007–2013. While changes in temperature throughout the year are substantial, their effect on the flux is negligible. As the surface area of the reservoir is assumed to be constant, the main change of the mean monthly fluxes is caused by variations in wind speed. As shown by the calculated fluxes at the highest hourly measured wind speeds (up to 25 m s^{-1}, see suppl. Fig. S6 and Table S9) over a period of 24 h, the mean monthly CO_2 flux can be increased by up to 50 %. The period of full ice cover is uncertain, but assuming a coverage of 3 months during winter would decrease the fluxes by around 25 %.

3.4.2. *Hálslón reservoir*

The data from the Hálslón reservoir show clearly that the pCO_{2w} values are always at or below the atmospheric pCO_{2atm} pressure (Figs. 3 and 4 and Table S9). All but the first two samples were sampled in August and September each year. Some samples are at less than 10 µatm. The measured depth profiles overall show a decrease of p_{CO2} with increasing depth. Caution must be taken, as the sample dates and methods were not uniform as described in the method and introduction sections. This is observed by very low pCO_{2w} values for one 5 m deep sample. The average pCO_{2w} of all the samples from the Hálslón reservoir was **174 µatm** (median: 138 µatm), while the corresponding average pCO_{2atm} during the study period of 2007–2013 was 392 µatm.

The samples collected from the Kárahnjúkar power plant outflow channel are mostly at or below atmospheric pCO_2 values as depicted by the red filled squares in Figs. 2 and 4 (Table S7). The highest measured value is at 814 µatm, approximately double the atmospheric pCO_2 value, some values are as low as the deepest samples from the Hálslón reservoir at ~10 µatm. Data interpretation of the outlet waters has to be done with caution, as the Kárahnjúkar hydropower station is fed by two additional reservoirs, that are partially mixing in the headrace tunnels with each other or with water from the Hálslón reservoirs in May to July each year.

Monthly mean fluxes for the Hálslón reservoir were calculated as described for Lagarfljót but multiplied with the average surface area (see Supplementary Information) for each month derived from the reservoir water level (see Fig. S1 and S2) and added up over the entire ice-free period. As the surface area in May is quite small (Fig. S1), the flux is very small too (Table S9), while the water fill level of the reservoir in November is nearly at its peak. This induces some additional uncertainty as the ice cover affects the gas flux. Therefore, the annual flux of the reservoir has been estimated by adding the values from June to November. Potential fluxes for May are provided in Fig. 3 and Table S9 but are not included in the annual flux.

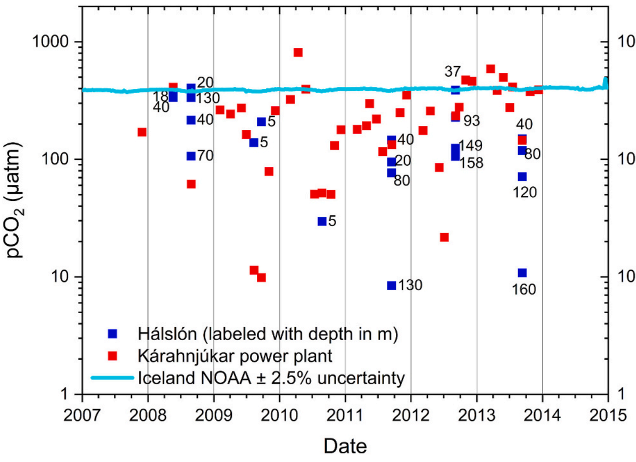

Fig. 4. Time evolution of the in situ partial pressure of CO_2 in the Hálslón reservoir water (blue filled squares) and in the outlet from the Kárahnjúkar power plant (red filled squares). The sample numbers refer to the Hálslón water depth (m) at which the samples were taken. The light blue curve represents the CO_2 partial pressure in the atmosphere at the NOAA Stórhöfði south Iceland CO_2 monitoring station (Lan et al., 2023). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The CO_2 flux from the Hálslón reservoir is estimated to be **−5323 ± 3100 t** over the entire ice-free period from June to November for each year. This value has a high uncertainty, as the average pCO_{2w} value of 174 µatm likely does not represent the overall reservoir. Changes in pCO_{2w} throughout the reservoir waterbody, as well as with seasons and over the years are large. Changes in the reservoir surface area contributed to water usage and precipitation increase the error. High wind speed events (Fig. S6 and Table S8) can lead to increased flux rates but are limited by ice cover and by the short event durations but can still be as large as 30 %.

4. Discussion

The highest calculated in situ partial pressure of CO_2 (pCO_{2w}) in the waters of the present study was ~850 µatm from the Lagarfljót reservoir before the construction of the Kárahnjúkar dam March 1st, 1999. This was more than twice the atmospheric pCO_{2atm} at the time of sampling (Fig. 2 and Table S4). Hence, CO_2 was then released from the water and to the atmosphere. Nevertheless, this is a relatively low maximum pCO_{2w} as values as high as 100,000 µatm are observed in organic-rich wetland soil waters before their exposure to the atmosphere (Linke et al., 2024a; Linke et al., 2024b). The lowest pCO_{2w} was about 10 µatm at 130–160 m depth within the Hálslón reservoir and comparable to the lowest value in the outlet from Kárahnjúkar power plant in late summer of 2009 (Figs. 2 and 4 and Table S6 and S7). The lowest reported pCO_{2w} in Iceland is around 0.32 µatm, measured in spring water emerging from glassy basaltic rocks at pH 10.10 and 2 °C (Gislason et al., 1996).

4.1. *Estimated CO_2 fluxes*

The total annual CO_2 flux from the Lagarfljót reservoir to the atmosphere was estimated to be around **5335 ± 2736 t** before the construction of Kárahnjúkar power plant. The annual flux decreased to one third, **1670 ± 2741 t,** for the observation period of 2007–2013. This is in direct correspondence with the change in ΔpCO_2 (pCO_{2w} - pCO_{2atm}) in eq. 1; from 140 µatm to 52 µatm, respectively and the effect on total (t yr^{-1}) and specific fluxes (t km^{-2} yr^{-1}) are the same at low windspeed (Fig. 5) as the geographic surface area of the Lagarfljót reservoir was the same during both periods. A 3-month ice cover during wintertime would decrease these fluxes further by ~25°%.

The CO_2 fluxes of the Hálslón reservoir are negative, from the atmosphere into the reservoir water, −5323 ± 3100 t over the entire ice-free period of around 6 months per year, making it a CO_2 sink. These total annual fluxes are calculated using average pCO_{2w} and average pCO_{2atm} values yielding a fixed CO_2 partial pressure gradient of −218 µatm. As described before, this is mostly based on samples collected in August–September each year and therefore shortly after the reservoir reached its maximum filling level and maximum geographical surface area. Hence, the August are most important for the total annual fluxes as shown in Fig. 3. These total fluxes are mainly affected by variation in ice cover, changes in total water surface area as a function of reservoir fill level and wind speed (Figs. S2 and S6). High wind speed events that lasted for only 24 h can affect the monthly mean flux by up to 30 %, but often occur during winter when the Hálslón reservoir is ice-covered. Apart from causing ice formation, temperature variations are less important, this can be seen in Lagarfljót (Fig. 3) over the months where

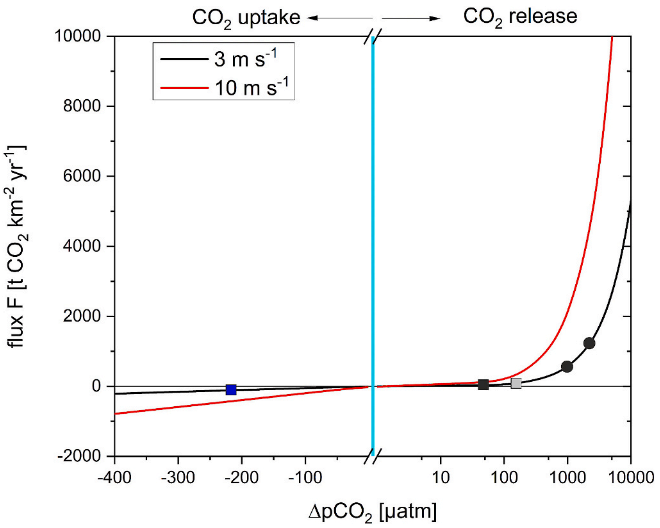

Fig. 5. The specific CO_2 flux F at 10 °C as a function of ΔpCO_2 (pCO_{2w} - pCO_{2atm}), assuming fixed wind velocities at 3.0 m s^{-1} and 10 m s^{-1}, the blue line at $\Delta pCO_2 = 0$ indicates the equilibrium between CO_2 partial pressure of the atmosphere and the water phase. Symbols represent values of the reservoirs from this study and the literature – Hálslón reservoir (blue square), Lagarfljót reservoir 1998–2003 (grey square), Lagarfljót reservoir 2007–2013 (black square), data from Saidi and Koschorreck, 2017 and Louis et al., 2000 as black circles. Note that CO_2 uptake at values below 0 is displayed in linear scale, while emissions are displayed in logarithmic scale to cover a larger scale. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the temperature varies by up to 10 °C but the wind speeds are relative constant, resulting in fluxes that are nearly equal throughout the year. Temperature changes of around 10 °C affect the fluxes within a few percentages, while as the increase of wind speed from 5 to 10 m s^{-1} results in more than doubling of the flux, and even more at higher wind speeds as the gas transfer velocity k_W becomes much larger with wind speed depending on the approach (see Supplementary Information and Fig. S6).

4.2. *Flux limitation by pCO_{2atm}*

While the specific emission of CO_2 oversaturated water bodies to the atmosphere can be large, such as those waters emerging from organic-rich wetland soils in southern Iceland (Linke et al., 2024a and b), the maximum potential partial pressure gradient (pCO_{2W} - pCO_{2atm}) and therefore the negative fluxes are limited by ΔpCO_2 (~400 μatm) as shown in Fig. 5. Assuming an atmospheric partial pressure pCO_{2atm} of roughly 400 μatm, the maximum uptake of CO_2 from the atmosphere at a pCO_{2W} value of zero is equal to ~800 t km^{-2} yr^{-1} at 10 °C and 10 m s^{-1} wind speed as shown in Fig. 5. Meanwhile, any positive ΔpCO_2 values that are greater than approximately 400 μatm ($pCO_{2W} > 800$ μatm) result in CO_2 emissions that are larger than any potential uptake under the before mentioned parameters, e.g. 2100 and 7800 t km^{-2} yr^{-1} for 3 m s^{-1} and 10 m s^{-1} respectively at a ΔpCO_{2W} value that is 10-times higher (4000 μatm).

4.3. *Comparison of total riverine fluxes and total water-air fluxes*

Eiriksdottir et al. (2017) calculated an increase of the dissolved inorganic carbon (DIC) flux from 19,016 t yr^{-1} to 34,048 t yr^{-1} for Lagarfljót at Lagarfoss (Fig. 1) and a decrease from 21,558 t yr^{-1} to 11,618 t yr^{-1} for Jökulsá á Dal at Hjardarhagi (Fig. 1) for the time periods 1998–2003 and 2008–2013 respectively, with a total increase of these DIC fluxes to the ocean by 13 %. These changes are attributed to the change of the river course following the damming of Jökulsá á Dal and the construction of the Kárahnjúkar power plant, and about 10 % climate induced increase in runoff during this period (Eiriksdottir et al., 2017). Compared to these annual river fluxes, the here presented potential annual CO_2 emissions from the Lagarfljót reservoir of 5335 t before and 1670 t after damming decreased to one third. Before the damming the annual CO_2 emissions from the Lagarfljót reservoir to the atmosphere were equal to 28 % of the riverine DIC fluxes to the ocean but decreased to 4.9 % after the damming. The CO_2 uptake by the Hálslón reservoir as a percentage of the Lagarfljót DIC flux is equal to 15 % for the time span 2008–2013.

4.4. *Role of photosynthesis and organic decay within these water bodies*

The Hálslón reservoir's average soil thickness before dam construction was 2.2 m and the carbon content was low: 10 % in the top part, 1.5–2.5 % in the middle and 3.8–4.8 % in the lower part of the soil sections (Arnalds and Gísladóttir, 2001). The bedrock consists of basaltic rocks that are relatively young, about 1 Myr (Gislason et al., 2009). The river suspended basaltic particles in Jökulsá á Dal at Brú, before damming, were reactive Mg-rich basalt (Eiriksdottir et al., 2008; Gislason et al., 2006), and as can be seen in Table S3, the average concentration of suspended particulate organic carbon particles was only 0.38 weight-% of the total river suspended inorganic particle concentration. Similarly, the average concentration of suspended organic carbon particles from the outlet of the Kárahnjúkar power plant was only 0.22 weight-% of the total concentration of suspended inorganic and organic particles in the river. Concentrations in Hálslón at various depths were 0.24 weight-%, highlighting the dominant effect of water rock interactions rather than organic processes such as photosynthesis, respiration, and break down of river transported organic matter.

Carbon to nitrogen (C:N) ratios of the minuscule river and reservoir suspended organic particulates are between 20 and 23 (median values) suggesting input from a terrestrial plant source (McGroddy et al., 2004). Lower C:N ratios close to 6.6 indicate algae or phytoplankton (Redfield 1934). In comparison, the C:N ratio of suspended organic particle matter in the outflow from lake Mývatn in NE-Iceland is close to 7 throughout most of the year (Eiríksdóttir et al., 2018). This lake is located approximately 100 km north-west of the Hálslón and Lagarfljót reservoirs. It is primarily fed by groundwater that interacts with basaltic rock. The lake is highly productive, mainly due to diatom production.

The C:N ratios of the Jökulsá á Dal river at Hjardarhagi increased from 23 to 29 (median values) following the change of the river course after the damming. This indicates that the contribution of the surrounding peat areas increased, as they are known to reach very high C:N ratios of 60 and above (Loisel et al., 2014). Meanwhile, the C:N ratios of the Lagarfljót reservoir also slightly increased from 20 to 22 (median values) following the construction of the power plant. This can be explained as the increased particle flux reduced the phytoplankton activity resulting in less photosynthesis. Simultaneously, the decreased primary production at Lagarfljót lead to less alkalinity generation, which is balanced by increased dissolution as the alkalinity values did not change following the damming. The C:N ratio of the Hálslón reservoir is much higher (28) than in Lagarfljót indicating much less phytoplankton activity as more particles are present. Therefore, the alkalinity in the Hálslón reservoir originates largely from the dissolution under the glacier and within the reservoir itself rather than from photosynthesis.

4.5. *Comparison of specific fluxes of the present with organic matter dominated reservoir fluxes*

The specific median flux of CO_2 to the atmosphere from the Lagarfljót reservoir over the entire year was estimated to be 100.7 ± 51.6 g_{CO2} m^{-2} yr^{-1} and 31.5 ± 51.7 g_{CO2} m^{-2} yr^{-1} before and after the damming respectively. These fluxes are relatively low compared to the specific fluxes from temperate reservoirs of 511 g_{CO2} m^{-2} yr^{-1} (21 reservoirs, Louis et al., 2000 and references therein) or German reservoirs of 611 g_{CO2} m^{-2} yr^{-1} (39 reservoirs, Saidi and Koschorreck, 2017), while data from tropical reservoirs typically show a higher average flux (e.g. 1277 g_{CO2} m^{-2} yr^{-1} in Louis et al., 2000, see Fig. 5). This is caused by much higher median pCO_{2w} values in tropical freshwater lakes 1910 μatm compared to non-tropical freshwater lakes 1120 μatm (Raymond et al., 2013). Therefore, our data shows that the annual CO_2 emissions from the Lagarfljót reservoir are much smaller compared to literature values of other reservoirs (see Fig. 5). The specific median flux of CO_2 from the atmosphere into the Hálslón reservoir, which was constantly undersaturated with respect to atmospheric pCO_{2atm}, was estimated to be -121.4 ± 67.9 g_{CO2} m^{-2} during the ice-free period of 6 months. If no ice cover was present the flux would be approximately -208 g_{CO2} m^{-2} yr^{-1}, with a large uncertainty caused by the varying geographical reservoir surface area and very high wind speeds observed during winter. The Hálslón reservoir exhibits much higher and reverse specific fluxes compared to the Lagarfljót reservoir, caused by a larger partial pressure gradient (ΔpCO_2). Specifically, the partial pressure gradients are 140 μatm for Lagarfljót (1998–2003), 47 μatm for Lagarfljót (2007–2013), and − 218 μatm for Hálslón (2008–2013). However, their total fluxes are similar. This is caused by the large surface area variation of the Hálslón reservoir over the year, and the presence of higher wind speeds at higher altitude compared to those at the protected Lagarfljót reservoir. The here observed fluxes are of the same order of magnitude as the estimated CO_2 drawdown by alkalinity export (~62 g_{CO2} m^{-2} yr^{-1}) from soil waters emerging from organic-rich soils in South Iceland or the average net annual carbon accumulation rate of corresponding soils (95–190 g_{CO2} m^{-2} yr^{-1}) (Linke et al., 2024b). Overall, the CO_2 sink effect that is observed in the Hálslón reservoir is small, which agrees with data from Barros et al. (2011) that observed on average only up to 4-times smaller influxes compared to reservoir emissions (see average emission values above). Additionally, CO_2 emissions from "young reservoirs" are often elevated in the first 15 to 20 years after their construction (Raymond et al., 2013), which is caused by the flooding and decomposition of organic material in soils. It is noteworthy that the soil present in the area of the Hálslón reservoir had a low carbon content (Gisladottir et al., 2014), hence flooding of this soil did not lead to large emissions.

The observed CO_2 drawdown is most likely caused by natural processes, as a result of mechanical and chemical weathering of the rocks at the base of the glaciers and airborne volcanic ash and dust (Gislason et al., 1996; Gislason et al., 2009; Eiriksdottir et al., 2013; Raiswell and Thomas, 1984) mostly under the glaciers before reaching the reservoirs. That the alkalinity originates from glacier-fed waters rather than reactions on-going within the Hálslón reservoirs can be seen when comparing alkalinity measurements from Hálslón reservoir. This is supported by the fact that the median alkalinity and pH of the river Jökulsá á Dal at Brú (see Supplement, Table S3) is close to the median alkalinity and pH of the outlet from the Kárahnjúkar power plant (Table S7). The dissolution of highly reactive basaltic particles within the catchment of the Hálslón reservoir and some within the water body of the reservoir creates alkalinity and raises the pH, resulting in CO_2 drawdown when in contact with the atmosphere (Gislason and Eugster, 1987a,b). The chemical weathering of the glacier fed rivers is highly affected by temperature and runoff (Gislason et al., 2009; Eiriksdottir et al., 2013) and will rise with increasing temperature and runoff and will be affected by changes in the river courses by damming.

5. Conclusions

Our work suggests that the diversion of water from one catchment to another caused by the construction of the Kárahnjúkar power plant led to a decrease in CO_2 emissions from the Lagarfljót reservoir over the study period. In comparison to the riverine DIC fluxes reported by Eiriksdottir et al. (2017), the CO_2 emissions to the atmosphere were only 28 % and 5 % of the total riverine DIC fluxes before and after the diversion of the rivers, respectively. The CO_2 uptake from the atmosphere of feeding reservoir Hálslón is about 5000 t CO_2 annually. The rate of CO_2 uptake is largely governed by the CO_2 partial pressure gradient (pCO_{2w} - pCO_{2atm}) across the air-water interface, windspeed, variation in geographic surface area, and sub-zero temperatures (ice formation). Although above zero degree Celsius temperature has less of an effect on the flux across the water-air interface, it will affect the water-rock interactions within the water body.

The here observed water rock interactions of glacier meltwater with basaltic bedrock, river suspended reactive material, and reactive dust produce considerable amounts of alkalinity. Newly created alkalinity leads to the uptake of CO_2 by the water bodies. These natural processes known for Icelandic glacier waters (e.g. Gislason et al., 1996) are similar to currently investigated enhanced rock weathering methods for CO_2 draw down from the atmosphere and can be considered as natural analogues, which can provide further insight (Linke et al., 2024a,b).

CRediT authorship contribution statement

T. Linke: Writing – review & editing, Writing – original draft, Visualization, Methodology. **E.S. Eiriksdottir:** Writing – review & editing, Investigation. **G.N. Petersen:** Writing – review & editing, Investigation. **S.R. Gislason:** Writing – review & editing, Writing – original draft, Supervision, Resources, Methodology, Funding acquisition, Conceptualization.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Tobias Linke reports financial support was provided by National Power Company of Iceland. Sigurdur Reynir Gislason reports financial support was provided by National Power Company of Iceland. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material associated with this article can be found in the online version. The Supplement contains additional explanations of the flux calculations and surface area estimates of the reservoirs, as well as associated tables and figures. The underlying numerical data of this work are provided in the supplementary tables. Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemgeo.2025.122662.

Data availability

All data are provided either in the manuscript or the associated supplementary information.

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